



Docket No: 19789-8

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*December 18, 2003.*

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**PATENT**

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**IN THE UNITED STATES PATENT & TRADEMARK OFFICE**

Applicant: van Ooij et al. :  
Serial No.: 09/356,926 : Group Art Unit: 1775  
Filed: July 19, 1999 : Examiner: Michael LaVilla  
For: **Silane Coatings For Metal** :

**DECLARATION UNDER 37 C.F.R. 1.132**

Mail Stop AF  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Dear Sir:

Dr. Wim J. van Ooij declares that:

1. He is a co-inventor of, and is familiar with the present U.S. Patent Application Serial No. 09/356,926, filed July 19, 1999, entitled "Silane Coatings for Metal." He is also familiar with the Official Actions issued in this application, including the Official Action dated July 18, 2003 and the prior art references cited therein.

2. The methods of treating a metal surface as defined by claim 1 of the present application are based upon the surprising discovery that the addition of one or more bis-silyl aminosilanes to a vinyl silane solution significantly improves the storage stability of the solution and significantly improves the corrosion protection provided by the solution. As demonstrated in the examples on pages 16-18 of the specification, untreated HDG panels

exhibited significant corrosion, while the addition of even a small amount of bis-(trimethoxysilylpropyl) amine (“A-1170”) to a vinyltrimethoxysilane solution provided astonishing improvements in corrosion protection.

3. In order to demonstrate that the claimed bis-silyl aminosilanes when combined with vinyl silane solutions provide unexpected results as compared to other multisilyl silane combined with vinyl silanes, the following additional experiments were carried out under his direction and control:

### **Experiment 1**

Silane solutions were prepared at 5% for vinyltriethoxysilane (VS) and bis-(trimethoxysilylpropyl) amine (“A-1170”). Additional silane solutions were prepared at 6% for VS and 1,2-bis-(triethoxysilyl) ethane (BTSE). A solvent (ethanol) was used to improve silane solubility. The solutions were stirred at room temperature in order to hydrolyze the silanes. Silane solutions were mixed for solutions 1-5 as described in Table 1. The pH of the resulting mixtures VS/BTSE (1-3) and VS/A-1170 (4-5) was adjusted, if needed, using acetic acid to 6. Alkaline cleaned hot-dip galvanized steel panels (HDG, available from ACT Laboratories) were treated using the VS/BTSE (1-3) and VS/A-1170 (4-5) mixtures and then cured at 100 °C for two hours.

The corrosion performance of the silane-treated HDG panels was evaluated using a salt spray test as described in the specification (ASTM-B117). The salt spray test results obtained after 24 hours are depicted in Figures 1-3. Figure 1 depicts the HDG panels treated with mixtures 1 and 2. Figure 2 depicts the HDG panels treated with mixtures 3 and 4. Figure 3 depicts the HDG panel treated with mixture 5. As can be seen from the Figures, mixtures 1-3 performed much better than silanes 4 and 5 after 24 hours of salt spray test. The former three only show edge corrosion with different degrees, while the later two depict heavy general corrosion all over the sample surface.

| Mixture ID | VS  | A-1170 | BTSE | Water | Ethanol |
|------------|-----|--------|------|-------|---------|
| 1          | 2   | --     | 4    | 38    | 56      |
| 2          | 2   | --     | 4    | 6     | 88      |
| 3          | 2   | --     | 4    | 20    | 74      |
| 4          | 4.5 | 0.5    | --   | 15    | 80      |
| 5          | 4.5 | 0.5    | --   | 5     | 90      |

TABLE 1

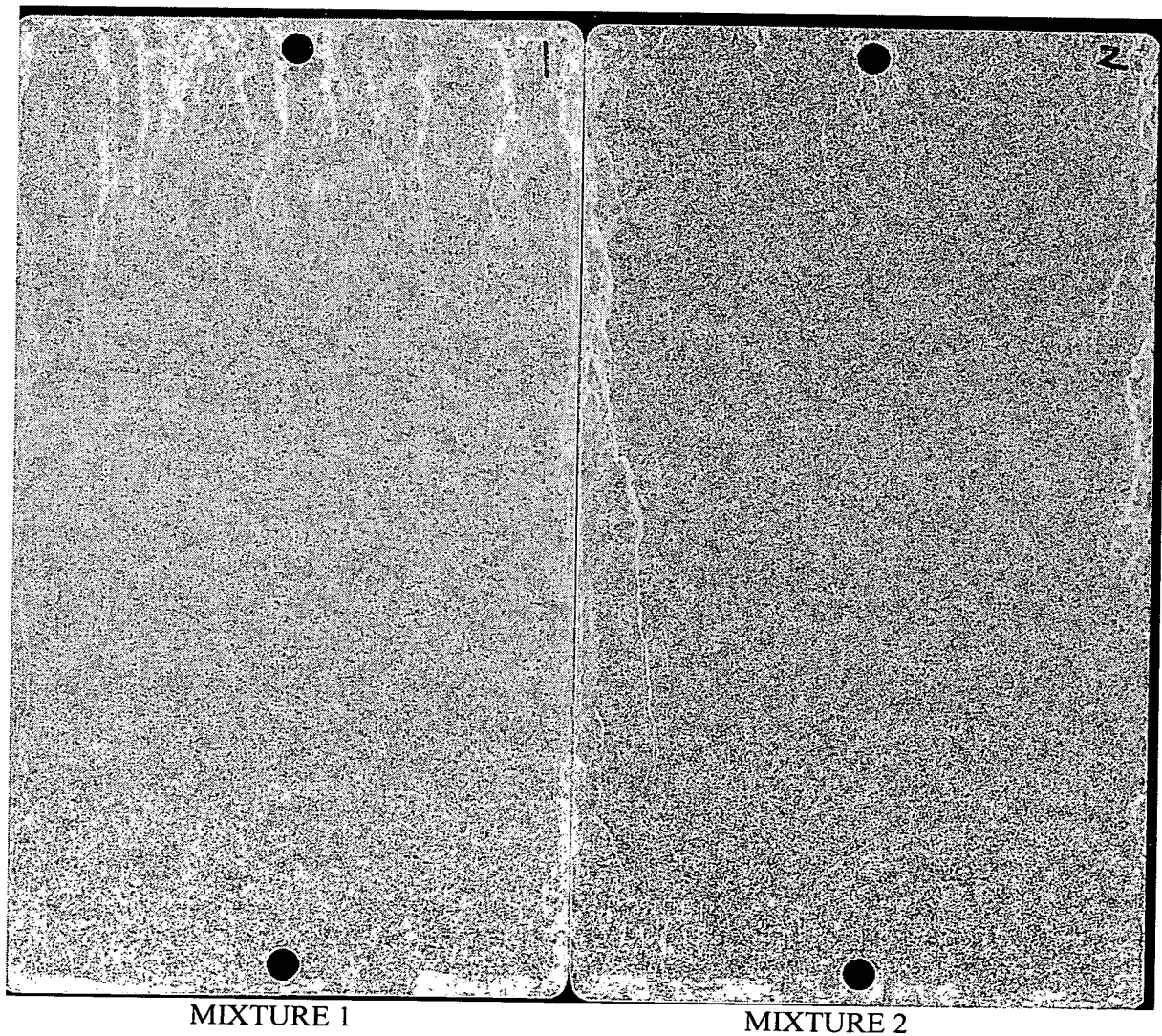
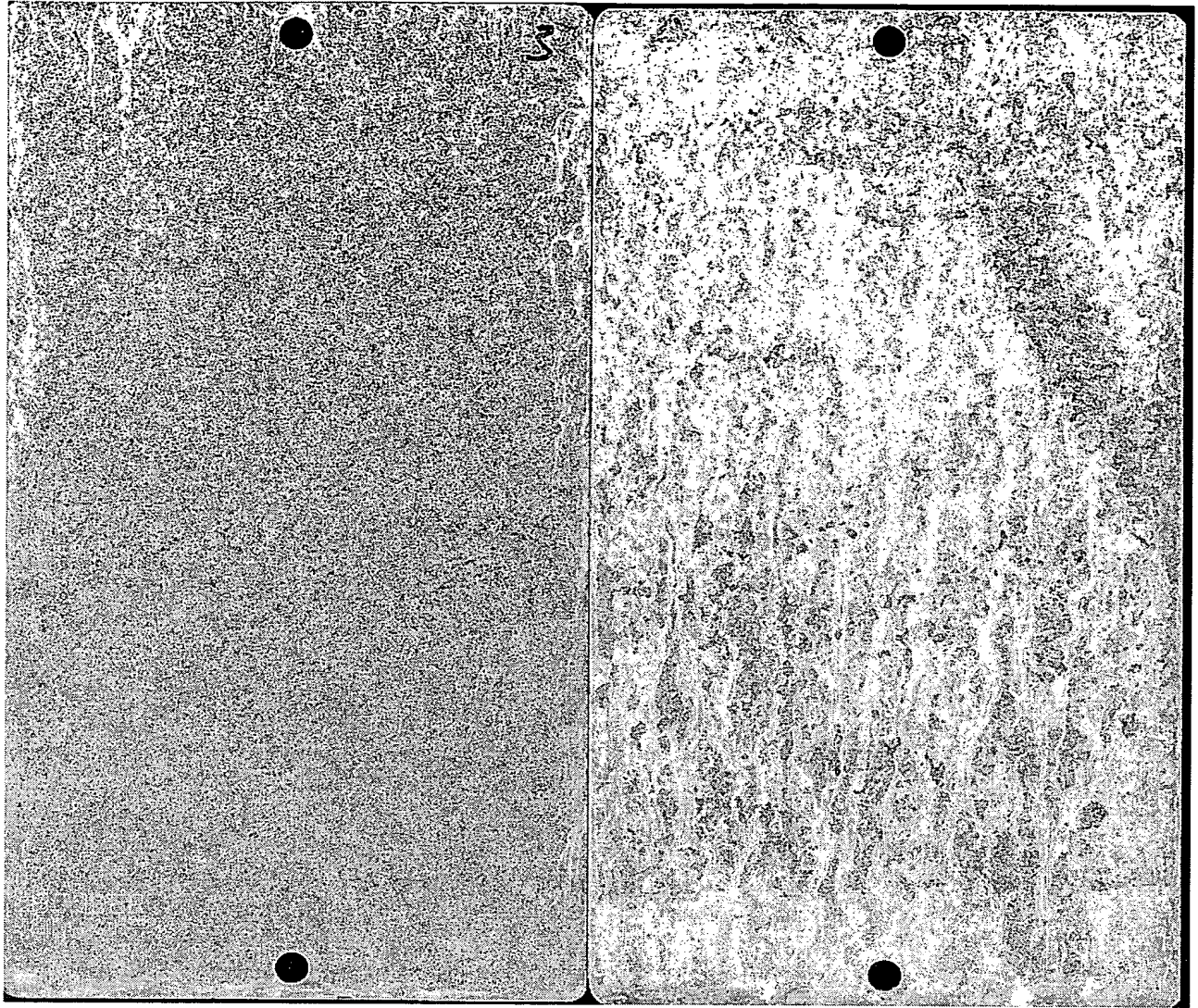


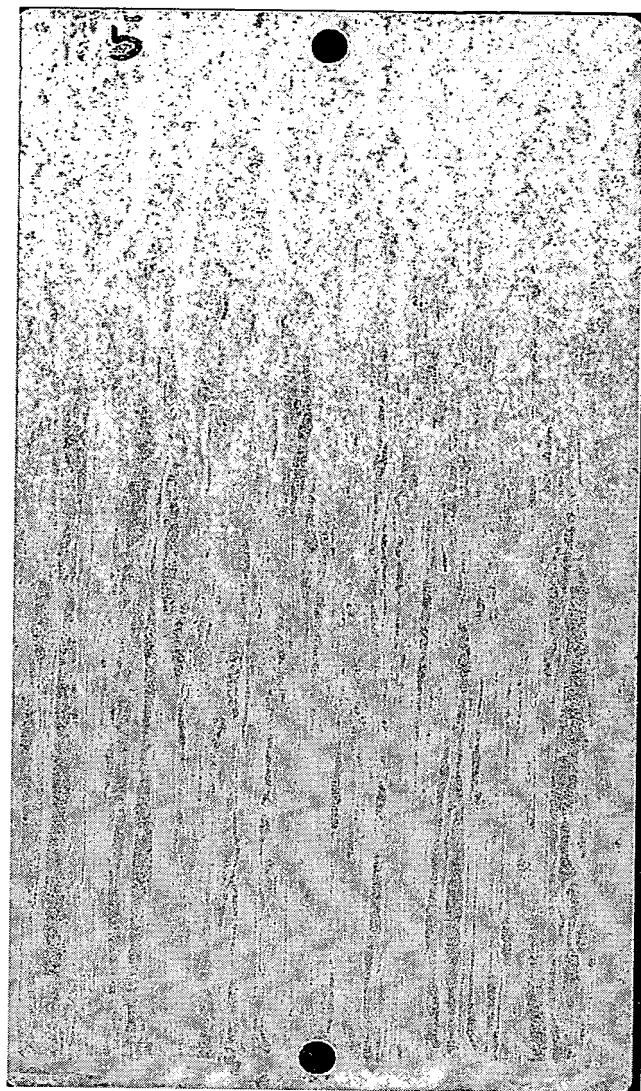
FIGURE 1



MIXTURE 3

FIGURE 2

MIXTURE 4



MIXTURE 5  
FIGURE 3

### **Experiment 2**

Silane solutions were prepared at 5% for vinyltriethoxysilane (VS) and bis-(trimethoxysilylpropyl) amine ("A-1170"). Additional silane solutions were prepared at 6% for VS and 1,2-bis-(triethoxysilyl) ethane (BTSE). A solvent (ethanol) was used to improve silane solubility. The solutions were stirred at room temperature in order to hydrolyze the silanes. Silane solutions were mixed for solutions 6-9 as described in Table 2. The pH of resulting mixture "6" was 8.5. The pH of the resulting mixtures "7" and "8" was adjusted using acetic acid to 6.5. The pH of resulting mixture "9" was 6. Alkaline cleaned hot-dip

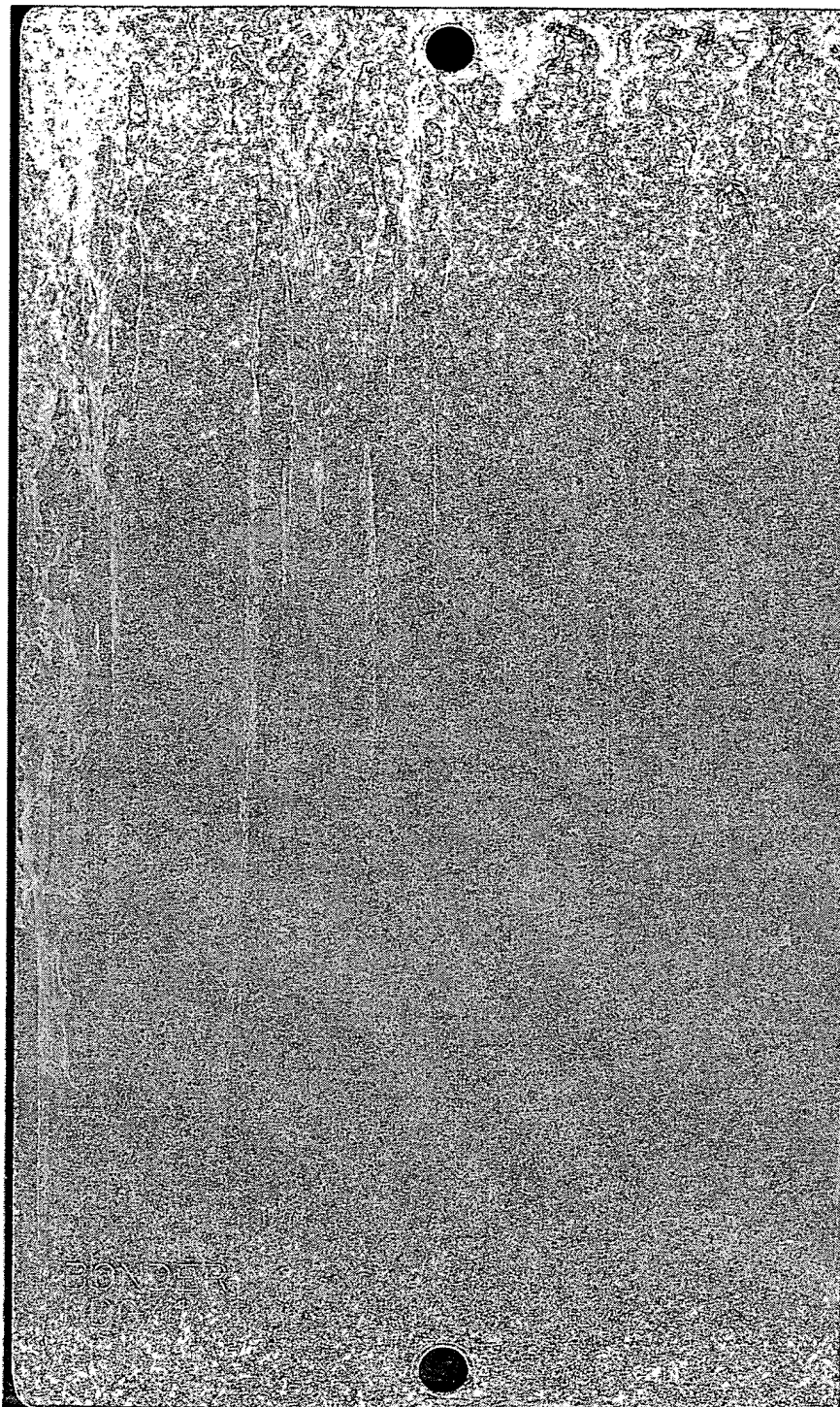
galvanized steel panels (HDG, available from ACT Laboratories) were treated using the VS/A-1170 (6-8) and VS/BTSE (9) mixtures and then cured at 120 °C for 5 minutes.

The corrosion performance of the silane-treated HDG panels were evaluated using a salt spray test as described in the specification (ASTM-B117). The salt spray test results are depicted in Figures 4-7. Figure 4 depicts the HDG panel treated with mixture 6. Figure 5 depicts the HDG panel treated with mixture 7. Figure 6 depicts the HDG panel treated with mixture 8. Figure 7 depicts the HDG panel treated with mixture 9. As can be seen from the Figures, mixtures 6-9 performed comparably in the 24 hour salt spray test.

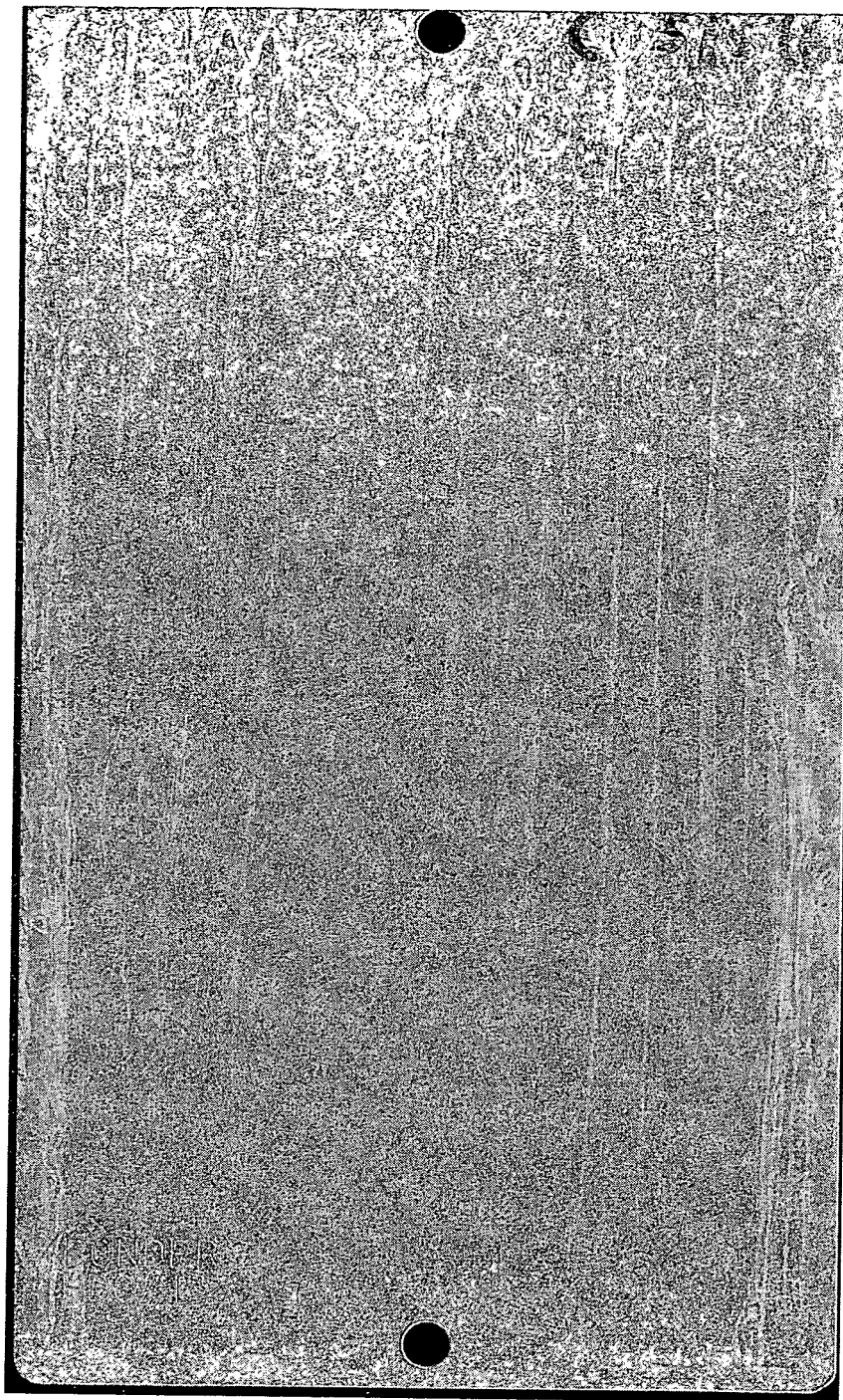
| Mixture ID | VS  | A-1170 | BTSE | Water | Ethanol |
|------------|-----|--------|------|-------|---------|
| 6          | 4.5 | 0.5    | --   | 5     | 90      |
| 7          | 4.5 | 0.5    | --   | 5     | 90      |
| 8          | 4.5 | 0.5    | --   | 15    | 80      |
| 9          | 2.0 | --     | 4.0  | 20    | 74      |

TABLE 2



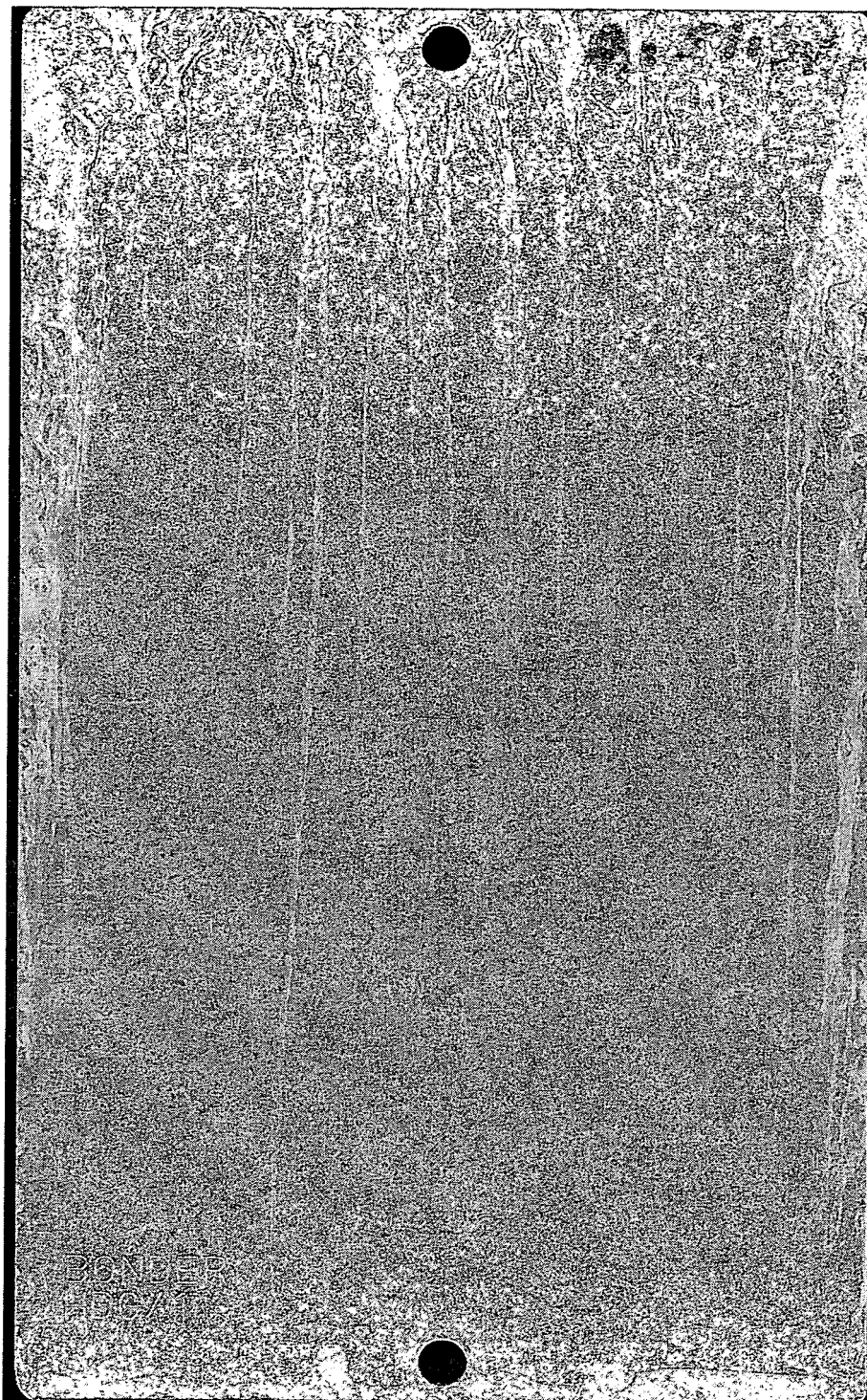


MIXTURE 6  
FIGURE 4

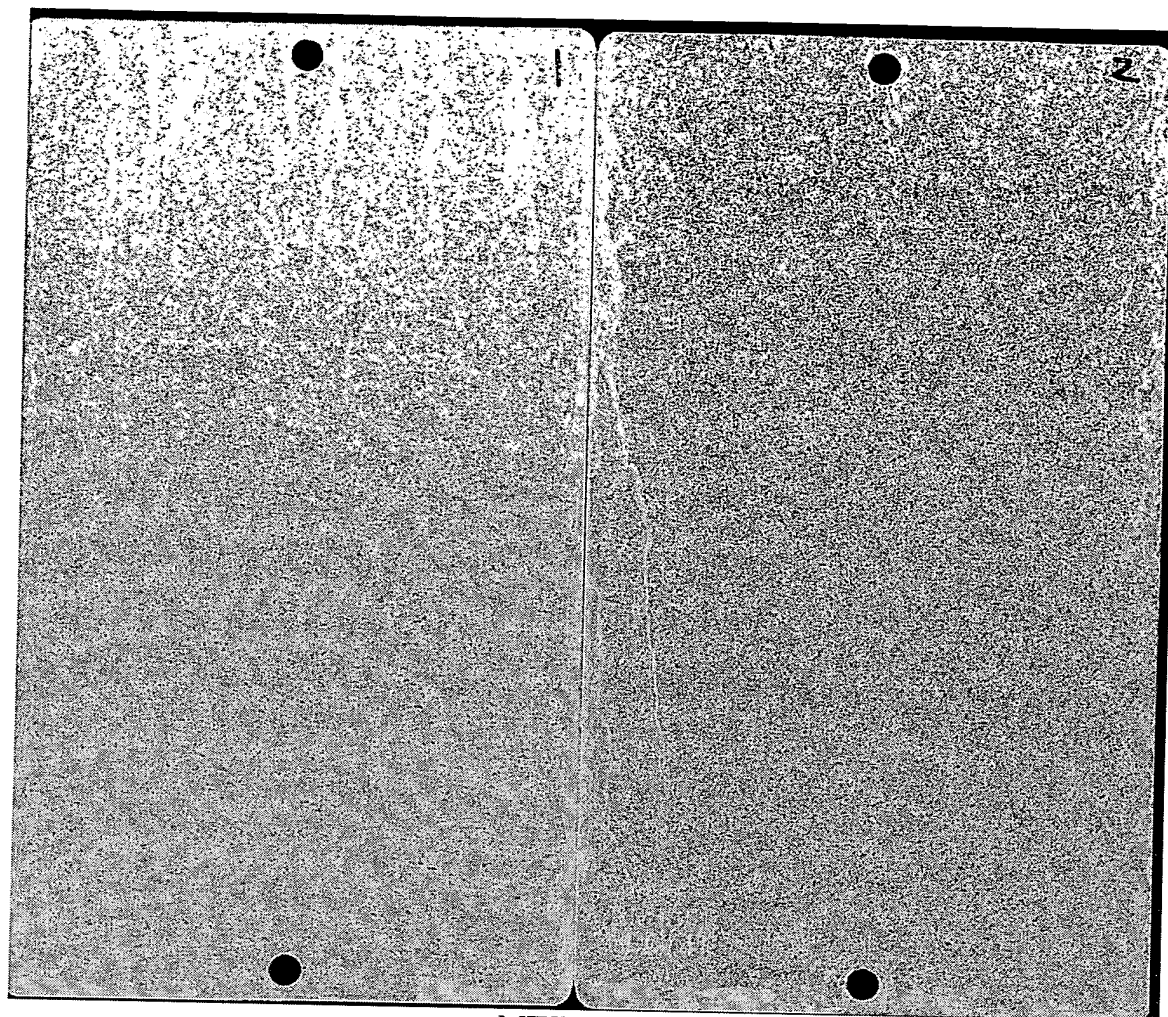


MIXTURE 7  
FIGURE 5





MIXTURE 8  
FIGURE 6



MIXTURE 9  
FIGURE 7

### **Experiment 3**

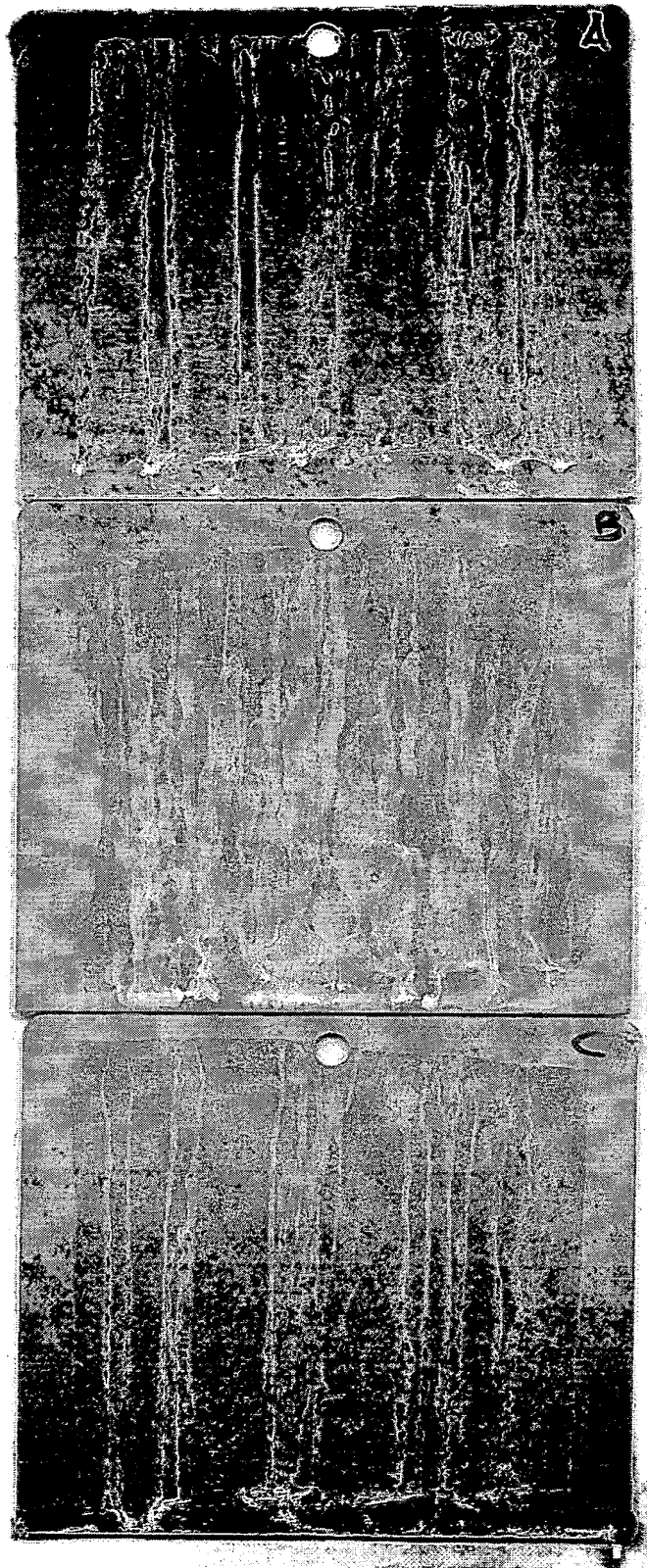
Silane solutions were prepared at 6% for vinyltriethoxysilane (VS), bis-(trimethoxysilylpropyl) amine ("A-1170") and 1,2-bis-(triethoxysilyl) ethane (BTSE). A solvent (ethanol) was used to improve silane solubility. The solutions were stirred at room temperature in order to hydrolyze the silanes. Silane solutions were mixed for solutions A-C as described in Table 3. The pH of resulting mixture A was 6. The pH of the resulting mixtures B and C were adjusted using acetic acid to 4. Alkaline cleaned hot-dip galvanized

steel panels (HDG, available from ACT Laboratories) were treated using the VS/BTSE (A and B) and VS/A-1170 (C) mixtures and then cured at 120 °C for 5 minutes.

The corrosion performance of the silane-treated HDG panels were evaluated using a salt spray test as described in the specification (ASTM-B117). The salt spray test results are depicted in Figures 8. As can be seen from Figure 8, mixtures C performed better than mixture A and much better than mixture B in the 24 hour salt spray test.

| Mixture ID | VS | A-1170 | BTSE | Water | Ethanol |
|------------|----|--------|------|-------|---------|
| A          | 4  | --     | 1    | 87    | 8       |
| B          | 2  | --     | 4    | 70    | 24      |
| C          | 2  | 4      | --   | 70    | 24      |

TABLE 3



MIXTURES A-C  
FIGURE 8

4. The results of Experiment 1 are believed to be inaccurate due to a number of potential factors. These factors include the possible contamination of the chemicals due to age; the HDG panels were cured at a lower temperature, 100 °C, than utilized in the examples in the present specification and the HDG panels lacked edge control. The lower temperature may have lead to ineffective curing of the silane mixtures on the HDG panels. As such, Experiment 2 was conducted with fresh chemicals and the same curing conditions as utilized in the present specification, 120 °C for 5 minutes. Mixtures 7 and 8 of Experiment 2 are nearly identical in composition to mixtures 4 and 5 from Experiment 1. A comparison of the HDG panels supports the conclusion that one or more of the above factors in Experiment 1 resulted in inaccurate data being generated.

5. Experiment 3 details the unexpected results of the claimed bis-silyl aminosilanes when combined with vinyl silane solutions as having substantial improvement in corrosion protection when compared to other multisilyl silanes combined with vinyl silane. The surprising improvements exhibited by the present compositions are not disclosed or rendered obvious by the prior art references cited in the Official Action dated July 18, 2003.

6. As one skilled in the art will appreciate, the showings of the unexpected results of the present experiments and the examples detailed in the present specification, i.e. vinyl silane mixtures containing bis-(trimethoxysilylpropyl) ethylene diamine or bis-(trimethoxysilylpropyl) amine demonstrate the unexpected results as compared to other multisilyl silanes combined with vinyl silanes.

7. Dr. Wim J. van Ooij further declares that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under



Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Respectfully submitted,



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Dr. Wim J. van Ooij

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18 December 2003

date

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